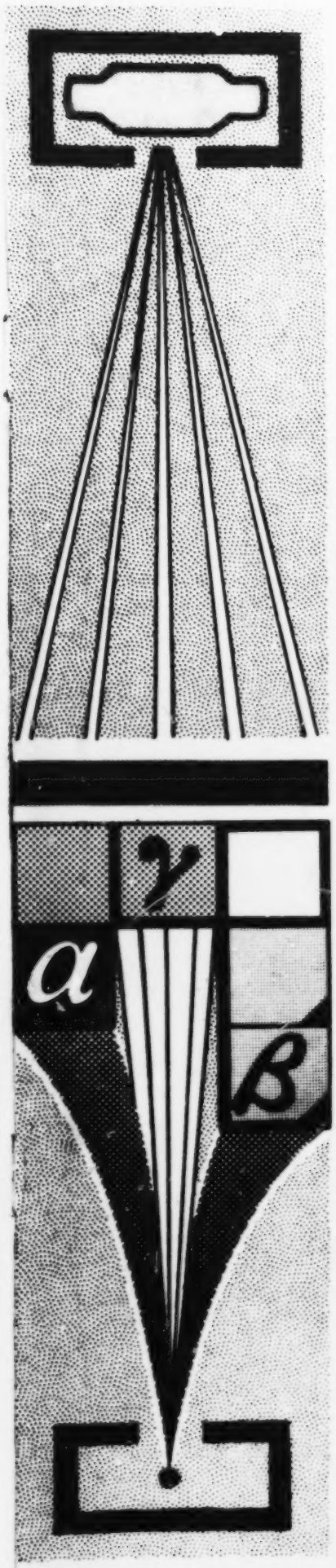

BRH/DMRE 70-4

PB196162

CYCLOTRON PRODUCTION of ^{123}I- an EVALUATION of the NUCLEAR REACTIONS WHICH PRODUCE THIS ISOTOPE

U.S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE
Public Health Service

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CYCLOTRON PRODUCTION of ^{123}I- an EVALUATION of the NUCLEAR REACTIONS WHICH PRODUCE THIS ISOTOPE

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OCTOBER 1970

U.S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE
Public Health Service

Bureau of Radiological Health
Rockville, Maryland 20852

FOREWORD

The Bureau of Radiological Health implements a national program designed to reduce the exposure of man to hazardous ionizing and nonionizing radiation.

Within the Bureau, the Division of Medical Radiation Exposure deals with 1) the reduction of unproductive ionizing radiation exposure of patients, workers, and others exposed by the use of x-rays and other machine-produced ionizing radiation, radioactive materials and radiopharmaceuticals, and 2) the improvement of radiological "systems" and methodology in the healing arts.

The Bureau publishes its findings in Radiological Health Data and Reports (a monthly publication), Public Health Service numbered reports, appropriate scientific journals, and technical report series for the Bureau's divisions, offices, and laboratories.

The technical reports of the Division of Medical Radiation Exposure allow comprehensive and rapid publishing of the results of intramural and contractor projects. The reports are distributed to State and local radiological health program personnel, Bureau technical staff, Bureau advisory committee members, university radiation safety officers, libraries and information services, industry, hospitals, laboratories, schools, the press, and other interested individuals. These reports are also included in the collections of the Library of Congress and the National Technical Information Service (formerly the Clearinghouse for Federal Scientific and Technical Information).

I encourage the readers of these reports to inform the Bureau of any omissions or errors. Your additional comments or requests for further information are also solicited.

John C. Villforth
Director
Bureau of Radiological Health

PREFACE

This document was prepared by the Nuclear Medicine Laboratory of the Division of Medical Radiation Exposure. The purpose of this research was to establish and describe the use of the various nuclear reactions by which ^{123}I, a superior, low-radiation dose radiopharmaceutical, can be cyclotron-produced.

Herein are reported both the methods of directly producing ^{123}I and those which indirectly produce the radionuclide through the β^+ decay of its natural precursor, ^{123}Xe. It is impossible to separate ^{123}I from the troublesome radioiodine contaminants, notably ^{124}I, which occur in the direct method. Thus, it is much preferable to produce pure ^{123}I from ^{123}Xe, which is easily separated from the radioiodines.

It is hoped that the findings of this research will stimulate the use of ^{123}I. Among its desirable characteristics is the capability of reducing the patient dose in a thyroid uptake measurement to a very small percentage of that delivered by the more commonly used ^{131}I. Since ^{131}I has been found responsible for most of the absorbed tissue dose in nuclear medicine, general adoption of pure ^{123}I could have far-reaching effects in reducing the medical radiation dose to the population.

Arve H. Dahl
Acting Director
Division of Medical Radiation Exposure

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1. INTRODUCTION

1.1 BACKGROUND

This work describes cyclotron techniques for production of ^{123}I. The production of this isotope has been prompted by its recognition as the most ideal radioisotope of iodine for low-exposure in vivo measurements (*1*). It has been calculated, for example, that the ^{123}I radiation dose to a patient from a human thyroid uptake test is only a few percent of that received with the more commonly used ^{125}I and ^{131}I (*2*). This reduced dosage stems from the fact that ^{123}I emits far less charged-particle radiation than does ^{131}I, and its physical half-life is only 0.9 and 6.9 percent of that of ^{125}I and ^{131}I, respectively. In addition, Wagner and Emmons, who have introduced formulas to determine the ideal radiopharmaceutical as one with a physical half-life no longer than necessary to ensure maximal activities in the organ studied at the optimal observation time, have found ^{123}I to be "more ideal" than either ^{125}I or ^{131}I (*3*). Furthermore, the 159 keV gamma ray of ^{123}I approaches an optimal energy that is low enough to be efficiently counted with thin crystals and to allow use of high resolution collimators, but is high enough to obtain necessary tissue penetration. All these desirable characteristics make this nuclide in demand for nuclear medical applications.

Preliminary clinical results of the use of ^{123}I for in vivo thyroid measurements suggest that although this radionuclide is advantageous for low exposure measurements, the ^{124}I contamination (0.5 - 1.0 percent) prevents full use of its capabilities as a high resolution, low radiation dose scanning agent (*4,5*).

1.2 EXISTING METHODS

To date, most of the ^{123}I used in nuclear medical applications has been produced by two nuclear reactions. Beaver, Holley, Eldridge, and Hupf have produced it at the ORNL 86-inch cyclotron by the 15.5 MeV proton bombardment of isotopically enriched ^{122}Te, according to the reaction $^{123}Te(p,n)\ ^{123}I$ (*6-8*). Goolden, Glass, and Silvester report its production at the Medical Research Council Cyclotron by the ^{4}He bombardment of natural antimony according to the reaction $^{121}Sb(^{4}He,2n)\ ^{123}I$ (*9*). Both methods have resulted in yields of ^{123}I that were adequate for nuclear medical applications, but were contaminated with other iodine isotopes with half-lives longer than that of ^{123}I.

The purity of the ^{123}I produced by the $^{123}Te(p,n)\ ^{123}I$ method is limited by the ^{124}Te, ^{125}Te, ^{126}Te, and ^{130}Te contaminants in the ^{123}Te target. Upon bombardment with protons, these targets impurities result in ^{124}I, ^{125}I, ^{126}I, and ^{130}I. The highest purity at which ^{123}Te is available without extraordinary enrichment procedures (which are presently economically impractical) is approximately 79 percent. Using targets of this purity, the contamination of ^{123}I with ^{124}I, ^{125}I, and ^{126}I has been reported to be about 0.9, 0.08, and 0.3 percent (*8*). The ^{130}I contamination is not reported.

The purity of the ^{123}I produced by the $^{121}Sb(^{4}He,2n)\ ^{123}I$ reaction on natural antimony is limited by two factors. First, the 42.8 percent ^{123}Sb in natural antimony results in ^{125}I via the $^{123}Sb(^{4}He,2n)\ ^{125}I$ reaction. Second, the energy of the alpha particle and the target thickness must be controlled so that the $(^{4}He,2n)$ reactions predominate because $(^{4}He,n)$ reactions on ^{121}Sb and ^{123}Sb produce ^{124}I and ^{126}I, respectively, and $(^{4}He,3n)$ reactions on ^{123}Sb produce ^{124}I.

Silvester, Sugden, and Watson have been able to control the energy of the ^{4}He beam on natural antimony targets so that the ^{124}I and ^{126}I content was kept down to 0.8 and 0.14 percent (*10*). They also obtained ^{125}I contamination resulting from ^{4}He reactions on the ^{123}Sb in the natural antimony at levels of 0.7 percent.

Myers and Anger have attempted to reduce the ^{125}I and ^{126}I contamination in ^{123}I by using targets of ^{121}Sb powder enriched to 99.4 percent (*1*). They report only slight contamination of their ^{123}I with other radioiodines, but do not give detailed results.

1.3 OBJECTIVES

The objective of the Nuclear Medicine Laboratory in this work was to promote the use of ^{123}I as a low-dose superior radiopharmaceutical. This was accomplished by studying the feasibility of various nuclear reactions that produce ^{123}I. Once produced, the ^{123}I was clinically evaluated and compared to the more commonly used ^{131}I as a diagnostic radiopharmaceutical.

Acceptability of methods for producing ^{123}I was based on the following considerations:

a. Availability and cost of target material.
b. Product yield and purity.
c. Ease of ^{123}I recovery from target.
d. Establishment of methods suitable for different types of cyclotrons.

1.4 PRESENT WORK

Studied in this work were the $^{121}Sb(^{4}He,2n)\ ^{123}I$, $^{121}Sb(^{3}He,n)\ ^{123}I$, $^{122}Te(d,n)\ ^{123}I$, $^{122}Te(^{4}He,p2n)\ ^{123}I$, $^{122}Te(^{3}He,pn)\ ^{123}I$ and $^{123}Te(^{3}He,p2n)\ ^{123}I$ reactions that produce ^{123}I directly and the $^{122}Te(^{4}He,3n)\ ^{123}Xe$, $^{122}Te(^{3}He,2n)\ ^{123}Xe$ and the $^{123}Te(^{3}He,3n)\ ^{123}Xe$ reactions that produce ^{123}I indirectly through the positron decay of 2.1 hour ^{123}Xe.

2. THEORY

2.1 NUCLEAR REACTIONS THAT PRODUCE ^{123}I

Several nuclear reactions using stable xenon, tellurium or antimony isotopes as target nuclei result in ^{123}I as a residual nucleus either directly or indirectly through its natural precursor, ^{123}Xe. A compilation of some of these reactions is listed in tables 1a and 1b. Only those reactions that are induced by bombardment with commonly available n, p, d, ^{3}He or ^{4}He sources are included.

Table 1a. Coulomb barrier and E_{BP} of nuclear reactions that produce ^{123}I directly

Reaction	Coulomb barrier (MeV)	E_{BP} (MeV)
$^{124}Xe(n,pn)\ ^{123}I$	9	14
$^{126}Xe(p,^{4}He)\ ^{123}I$	16	6
$^{120}Te(^{4}He,p)\ ^{123}I$	15	16
$^{122}Te(p,\gamma)\ ^{123}I$	8	-3
$^{122}Te(d,n)\ ^{123}I$	8	7
$^{122}Te(^{3}He,pn)\ ^{123}I$	16	13
$^{122}Te(^{4}He,p2n)\ ^{123}I$	15	34
$^{123}Te(p,n)\ ^{123}I$	8	12
$^{123}Te(d,2n)\ ^{123}I$	8	14
$^{123}Te(^{3}He,p2n)\ ^{123}I$	16	20
$^{124}Te(p,2n)\ ^{123}I$	8	21
$^{124}Te(d,3n)\ ^{123}I$	8	25
$^{125}Te(p,3n)\ ^{123}I$	8	28
$^{121}Sb(^{3}He,n)\ ^{123}I$	15	5
$^{121}Sb(^{4}He,2n)\ ^{123}I$	15	25
$^{123}Sb(^{3}He,3n)\ ^{123}I$	15	20
$^{123}Sb(^{4}He,4n)\ ^{123}I$	15	41

Table 1b. Coulomb barrier and E_{BP} of nuclear reactions that produce ^{123}I from positron decay of 2.1 H ^{123}Xe

Reaction	Coulomb barrier (MeV)	E_{BP} (MeV)
$^{124}Xe(n,2n)\ ^{123}Xe$	0	18
$^{120}Te(^{4}He,n)\ ^{123}Xe$	15	16
$^{122}Te(^{3}He,2n)\ ^{123}Xe$	16	14
$^{122}Te(^{4}He,3n)\ ^{123}Xe$	15	35
$^{123}Te(^{3}He,3n)\ ^{123}Xe$	16	21

2.2 FEASIBILITY OF REACTIONS

2.2.1 Availability and Cost of Target Materials

Listed in table 2 are the naturally occurring nuclides that are potential target materials for ^{123}I production. Tabulated for each nuclide are its natural abundance and availability and cost in the isotopically enriched form. Because isotopic impurities in the target material usually result in radioiodine contaminants, notably ^{124}I, ^{125}I, ^{126}I, and ^{131}I, the most highly enriched target material is desirable for pure ^{123}I production. The ^{121}Sb, ^{123}Sb, ^{122}Te, ^{124}Te, and ^{125}Te are all available at the >90 percent enrichment level at a cost of a few dollars per milligram. The ^{123}Te is available at the 65-80 percent enrichment level at a cost of $13.50/mg which may be considered expensive in some instances. The ^{120}Te, at a cost of $69 for the 30-50 percent enriched material and ^{124}Xe and ^{126}Xe at a cost of $24.35/ml (S.T.P.) for the 10 and 4 percent enriched material, respectively, are prohibitively expensive for routine ^{123}I production. Consequently, only ^{121}Sb, ^{123}Sb, ^{122}Te, ^{123}Te, ^{124}Te, and ^{125}Te were considered suitable target materials for experimentation.

2.2.2 Reaction Cross Sections

Listed in tables 1a and 1b are the calculated coulomb barriers and the estimated energy E_{BP} of the bombarding particle at which the theoretical cross section for each reaction is assumed to be maximum. The coulomb barrier is the electrostatic potential energy that opposes interaction between a charged particle and a nucleus (*11*).

If the kinetic energy of the bombarding particle does not exceed the coulomb barrier, there is a low probability (cross section) that a reaction will occur. The E_{BP} for each reaction was estimated assuming that the maximal cross section of the desired reaction (a,b) occurs at the threshold for the (a,b+n) reaction (*12*). For example, the cross section of the $^{121}Sb(^{4}He,2n)\ ^{123}I$ reaction should be maximum at the threshold for the $^{121}Sb(^{4}He,3n)\ ^{122}I$ reaction.

Table 2. Availability and cost of potential target materials[a]

Nuclide	Natural abundance (percent)	Enriched abundance (percent)	Cost ($/mg)
^{121}Sb	57.25	95-99.5	2.15
^{123}Sb	42.75	95-99	2.50
^{120}Te	0.09	30-50	69.00
^{122}Te	2.46	90-95	3.60
^{123}Te	0.87	65-80	13.50
^{124}Te	4.61	85-95	1.90
^{125}Te	6.99	90-97	1.25
^{124}Xe	0.10	10	[b]24.35
^{126}Xe	0.09	4	[b]24.35

[a] Sb and Te enrichment and cost figures from ORNL, Isotopes Development Center, Oak Ridge, Tennessee. Xe from Mound Laboratory, Miamisburg, Ohio.

[b] $/ml at S.T.P.

A study of the coulomb barriers and E_{BP} for the reactions listed in tables 1a and 1b shows that very low cross sections could be expected for the $^{126}Xe(p,^{4}He)\ ^{123}I$, $^{122}Te(p,\gamma)\ ^{123}I$, and $^{121}Sb(^{3}He,n)\ ^{123}I$ reactions. The $^{122}Te(d,n)\ ^{123}I$, $^{122}Te(^{3}He,pn)\ ^{123}I$ and the $^{122}Te(^{3}He,2n)\ ^{123}Xe$ reactions would be considered borderline cases. Elimination of those reactions that have serious coulomb barrier problems and that are limited by target material considerations (see section 2.2.1) leaves the following feasible reactions:

Studied in this work

$^{122}Te(d,n)\ ^{123}I$
$^{122}Te(^{3}He,pn)\ ^{123}I$
$^{122}Te(^{4}He,p2n)\ ^{123}I$
$^{123}Te(^{3}He,p2n)\ ^{123}I$
$^{121}Sb(^{3}He,n)\ ^{123}I$
$^{121}Sb(^{4}He,2n)\ ^{123}I$
$^{122}Te(^{3}He,2n)\ ^{123}Xe$
$^{123}Te(^{3}He,3n)\ ^{123}Xe$
$^{122}Te(^{4}He,3n)\ ^{123}Xe$

Studied elsewhere

$^{123}Te(p,n)\ ^{123}I$

Not yet studied

$^{123}Te(d,2n)\ ^{123}I$
$^{124}Te(p,2n)\ ^{123}I$
$^{124}Te(d,3n)\ ^{123}I$
$^{125}Te(p,3n)\ ^{123}I$
$^{123}Sb(^{3}He,3n)\ ^{123}I$
$^{123}Sb(^{4}He,4n)\ ^{123}I$

All of the above mentioned reactions are within the energy limitations of nuclear research facilities. However, special emphasis was given to those reactions that are within the energy range of the "compact" cyclotron which is suitable for use by major nuclear medical facilities. The "small" or "compact" cyclotron is capable of accelerating 15-20 MeV protons, 7-10 MeV deuterons, 20-30 MeV ^{3}He and 15-20 MeV ^{4}He particles.

3. EXPERIMENTAL APPARATUS

3.1 Ge(Li) SPECTROMETER

The identification and assay of gamma emitting radionuclides were done using a Ge(Li) spectrometer. The spectrometer included a 6 cm^2 area x 0.7 cm deep Ge(Li) detector coupled to a low noise, charge sensitive, preamplifier-amplifier system and a 1600 channel pulse-height analyzer.

3.2 CYCLOTRONS

The cyclotrons used in this work were the following:

a. University of Indiana (IU) ^4He (22.5 MeV)
   Bloomington, Indiana
b. Lewis Research Center (LRC) d (21.0 MeV), ^4He(42.2 MeV)
   Cleveland, Ohio
c. Ohio State University (OSU) ^3He (18 MeV)
   Columbus, Ohio
d. Argonne National Laboratory (ANL) ^3He (35.5 MeV)
   Argonne, Illinois
e. Sloan Kettering Institute (SKL) ^3He (23.0 MeV)
   New York

3.3 STANDARD TARGET

Figure 1 shows the diagram of the target holder used at the LRC cyclotron where most of the bombardments were done. Other cyclotrons used targets similar in principle but different in design.

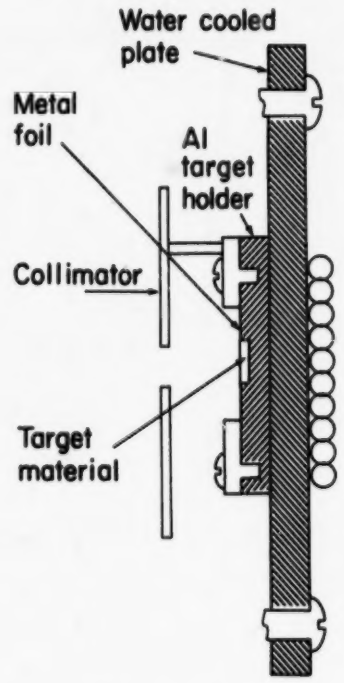

Figure 1. LRC cyclotron target holder

The metallic target powder was packed into the 3/8-inch cavity milled into the aluminum target plate and covered with an absorber foil. The area of the cavity was slightly larger than that of the aperture in the carbon collimator so that only that portion of the holder containing target material was irradiated. The volume of the cavity was just large enough to contain the compacted powder so that, when the plate was covered and in the vertical position, the powder distribution remained constant. The absorber foil prevented escape of target material into the cyclotron's vacuum system and also attenuated the beam energy to the desired value. The aluminum target holder was bolted to a water-cooled 6-inch copper plate which was bolted onto the end of a cyclotron beam tube. Irradiations were done in vacuum.

The area and position of the incident beam were determined before each bombardment by televiewing its fluorescence on a zinc sulfide window positioned on the beam tube in place of the target assembly. The beam was adjusted in area to a 0.5-inch square with roughing slits so that it completely covered the area of the target collimator. However, after some bombardments, it was obvious from the uneven discoloration of the covering foil and the condition of the target powder that the target was not hit uniformly.

3.4 GAS FLOW TARGET ASSEMBLY

Figure 2 shows a general diagram of a continuous gas flow generator.

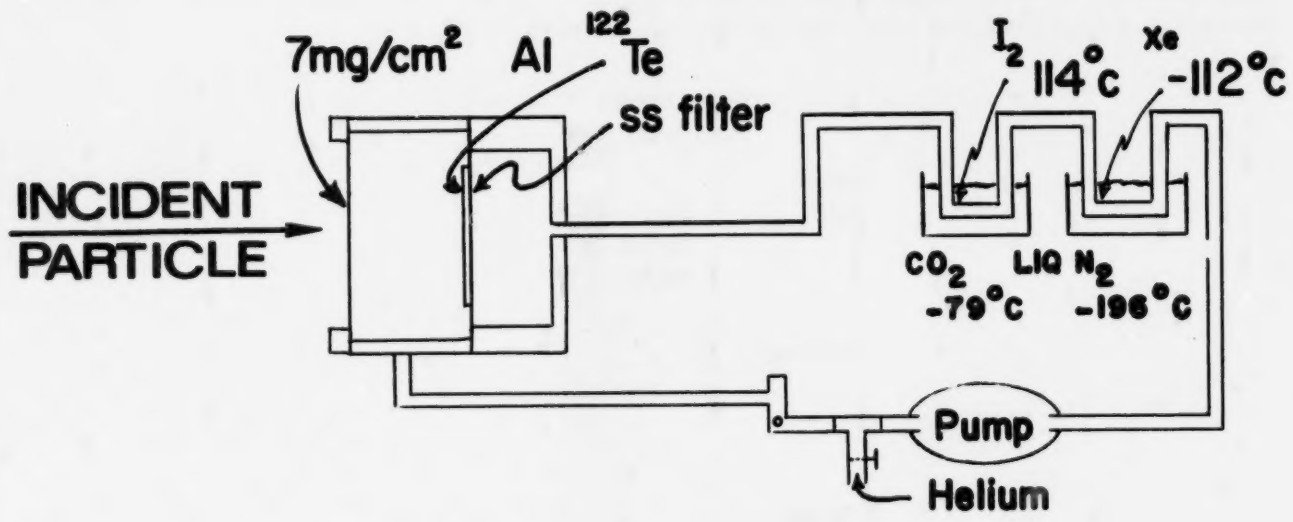

Figure 2. Continuous flow ^{123}I generator

The target assembly consisted of a water-cooled aluminum cylinder separated into two chambers by a stainless steel screen which supported the target material. The chamber to the right of the target was connected with Tygon tubing to a 1/4-inch copper coil trap which was maintained at a temperature of -79°C in a Dewar of CO_2. The CO_2 trap was connected to another 1/4-inch copper, tygon or glass trap maintained at a temperature of -196°C in a Dewar of liquid nitrogen. The liquid nitrogen trap was then connected to a self-contained gas pump, a series of stopcocks, a flow meter and finally back into the target chamber to the left of the filter.

With the target chamber in the horizontal position, i.e., at 90° to that shown in the diagram, the tellurium powder was spread uniformly on the filter and the chamber covered with a 7 mg/cm^2 aluminum window. The generator was purged with helium gas through a series of stopcocks at a flow rate of about 10 l/min. After purging, the generator was sealed off and the flow rate maintained with the pump. With the flow rate maintained, the target chamber was then turned vertically as shown in the figure and inserted into the horizontal alpha particle beam of the cyclotron; the helium flow held the tellurium powder in place on the filter. During the bombardment, the volatile products were swept out of the target by the recirculating flow of helium gas into the CO_2 trap. At this trap, directly produced radioiodine and other contaminants with freezing points higher than -79°C were frozen out. The helium flow was then directed into the liquid nitrogen trap where ^{123}Xe produced from the desired reaction and other products with freezing points between -79°C and -196°C were isolated. The helium, which was still in the gaseous state, was recirculated through the target system.

Two variations of the above method were also used. The single pass method employed the above apparatus with a non-recirculating flow of helium gas. With the extremely slow flow rate of 25 ml/min, 90 percent of the released ^{123}Xe was isolated in the liquid nitrogen trap with very little expenditure of helium. At this slow helium flow rate, the solid CO_2 trap was very effective in eliminating directly formed radioiodine contaminants.

The cyclic method involved irradiating the tellurium powder in a sealed target holder, after which the tellurium is heated to drive off the ^{123}Xe and radioiodine contaminants. A trapping system identical to that described above is used to purify and isolate the ^{123}Xe. This cyclic method allowed only short bombardments, ~2 hours since the decaying ^{123}Xe is not being continuously removed from the target.

4. CALIBRATION

4.1 RESOLUTION, ENERGY CALIBRATION, AND LINEARITY

The resolution (FWHM) of the spectrometer for the 661.6 keV gamma ray of ^{137}Cs was 3.2 keV. Energy calibration was done with ^{57}Co (122.0 keV), ^{137}Cs (661.6 keV) and ^{60}Co (1173.2 and 1332.5 keV). Radioiodine identification was accomplished by both energy and half-life analysis. Linearity of the detection system was checked by comparing the pulse height ratio to the gamma ray energy ratio. Gamma rays of ^{57}Co, ^{131}I and ^{137}Cs were used. The difference between the pulse height ratio and the energy ratio for ^{57}Co and ^{131}I, i.e., between 121.98 and 364.47 keV, was less than 0.02 percent of the average ratio; that between ^{131}I and ^{137}Cs, i.e., between 364.47 and 661.64 keV, was 0.12 percent.

4.2 SPECTROMETER EFFICIENCY

The counting efficiency of the spectrometer was determined for 1 ml liquid samples in plastic vials centered directly on the detector. The energies and abundances of the gamma rays of standard radionuclides and of the radioiodines used in the assay calculations are listed in table 3.

Table 3. Energies (E_γ) and abundances (Ab_γ) of gamma rays of standard radionuclides and radioiodines[a]

Standard	E_γ (keV)	Ab_γ (percent)	Radio-iodine	E_γ (keV)	Ab_γ (percent)
^{125}I	35	7	^{125}I	35	7
^{141}Ce	145	48	^{123}I	159	83
^{139}Ce	165	80	^{131}I	364	82
^{131}I	364	82	^{126}I	388	34
^{134}Cs	605	98	^{130}I	418	35
^{137}Cs - ^{137m}Ba	662	85	^{124}I	602	67

[a]From Table of Isotopes, 6th Ed., Lederer, C. M., Hollander, J. M. and Perlman, I., John Wiley and Sons, Inc., New York (1967).

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5. DATA TREATMENT

5.1 SPECTRAL ANALYSIS

The counting rate of a given radioiodine was determined by computer integration under its respective gamma-ray photopeak. The computer integration program subtracted the Compton continuum (i.e., a "valley to valley" background) from each photopeak. This same procedure was used in determining the efficiency of the spectrometer for gamma-ray emitting standards.

The half-lives of radionuclides were determined by a least squares analysis of counting rates obtained at various times after bombardment.

The raw data was read out of the spectrometer on ASCll Teletype tape which was fed into an IBM 1130 computer. The computer provided photopeak integration and a 1600 channel spectrum via a Cal Comp Plotter. The photopeak integration data from decaying sources could be transferred to IBM cards and analyzed with another computer program to provide half-life information.

5.2 ERRORS

For the most part, the errors in the experiment resulted from counting statistics and uncertainties in spectrometer efficiency and irradiation parameters.

The counting errors at the 95 percent confidence level for ^{123}I and ^{124}I were less than 1 percent; for ^{125}I, ^{126}I, ^{130}I, and ^{131}I were normally less than 5 percent. In a few instances, when the counting rates of small quantities of contaminants were low, the errors were as high as $\pm$25 percent.

The errors in spectrometer efficiency were estimated to be less than $\pm$3 percent.

The greatest errors in the experiment were due to uncertainties in the irradiation parameters. The uncertainties resulted from nonuniformities in the incident beam density, unevenness of the target thickness, misalignment of the incident beam over the entire area of the target and incomplete transfer of target powder from the target holder to the distillation apparatus for radioiodine separation. The magnitude of these errors can be approximated from a statistical analysis of results from similar bombardments. The arithmetic mean and its standard deviation were calculated from the results of 12 similar alpha particle bombardments of ^{121}Sb (see section 6.1.2, table 5). The average ^{123}I yield was 0.60 $\pm$ 0.18 mCi/μA-h; the average ^{124}I, ^{125}I, and ^{126}I contaminants were 0.79, $\pm$0.13, (1.5 $\pm$ 0.2) x 10^{-2} percent and (2.6 $\pm$ 0.4) x 10^{-3} percent, respectively.

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6. EXPERIMENTAL METHOD AND RESULTS

6.1 $^{121}Sb(^{4}He,2n)\ ^{123}I$

6.1.1 Experimental Considerations

The E_{BP} for the $^{121}Sb(^{4}He,2n)\ ^{123}I$ reaction is 25 MeV which is well above the 15 MeV coulomb barrier. The 98.4 percent ^{121}Sb target powder was purchased from ORNL. The main impurity in this material was 1.6 percent ^{123}Sb. The radioiodine contaminants produced from this method are ^{125}I and ^{126}I from $^{123}Sb(^{4}He,2n)\ ^{125}I$ and $^{121}Sb(^{4}He,n)\ ^{126}I$ reactions and ^{124}I from $^{121}Sb(^{4}He,n)\ ^{124}I$ and $^{123}Sb(^{4}He,3n)\ ^{124}I$ reactions.

Because ^{125}I is produced from (^{4}He,2n) reactions, just as is the desired ^{123}I, its level of contamination is fixed by the ^{123}Sb impurity in the target material. Since ^{124}I and ^{126}I are produced from the competing (^{4}He,n) and (^{4}He,3n) reactions, their contamination levels can be minimized by using precise energy selection and extremely thin targets (10-15 mg/cm^2). However, when larger amounts of ^{123}I are desired, thicker targets (70 mg/cm^2) must be used. Because of the attenuation of beam energy in the thicker targets, incident energies higher than E_{BP} must be used. Furthermore, because (^{4}He,3n) reactions on ^{121}Sb produced 3.5 m ^{122}I, still higher incident energies may be desirable to reduce the (^{4}He,n) cross section which produces 4.2 d ^{124}I.

6.1.2 Results

Some typical Ge(Li) spectra of an iodine fraction that was distilled from an ^{121}Sb target are shown in figure 3. Spectra a-d of figure 3 were obtained at 22 hours, 6, 18, and 50 days, respectively, after the end of a bombardment of 98.4 percent enriched ^{121}Sb with 31 MeV alpha particles. Spectra b-d of figure 3 were plotted at factors of 10^1, 10^2, and 10^3, respectively, lower in relation to figure 3(a) to aid in visual comparison. No major activity was found above 800 keV.

Identified in figure 3(a) are the 27.5 and 31.0 keV x-rays and the 158.9, 248.1, 280.7, 346.3, 439.9, 505.1, 528.9, 538.5, 624.4, 687.9, 697.5, 735.9, and 783.0 keV gamma-ray photopeaks of ^{123}I. The photopeaks at 185.3 and 317 keV are random sums of abundant photons. The 185.3 keV peak is a sum of the 158.9 keV gamma-ray photon of ^{123}I and the x-ray photon of tellurium; the 317 keV peak is a sum of two 158.9 keV photons. With lower activity sources these sum peaks are not seen. Also identified in figure 3(a) are the 511.0 keV annihilation peak and the 602.4, 644, and 722.4 keV gamma-ray photopeaks of ^{124}I. The ^{124}I contamination was 0.70 percent at the end of this particular bombardment.

In figure 3(b) only 0.06 percent of the 13.3 hour ^{123}I and 37 percent of the 4.2 day ^{124}I remained in the sample as evidenced by the persistence of the 158.9 and 511.0, 602.4, 644, and 722.4 keV photopeaks, respectively.

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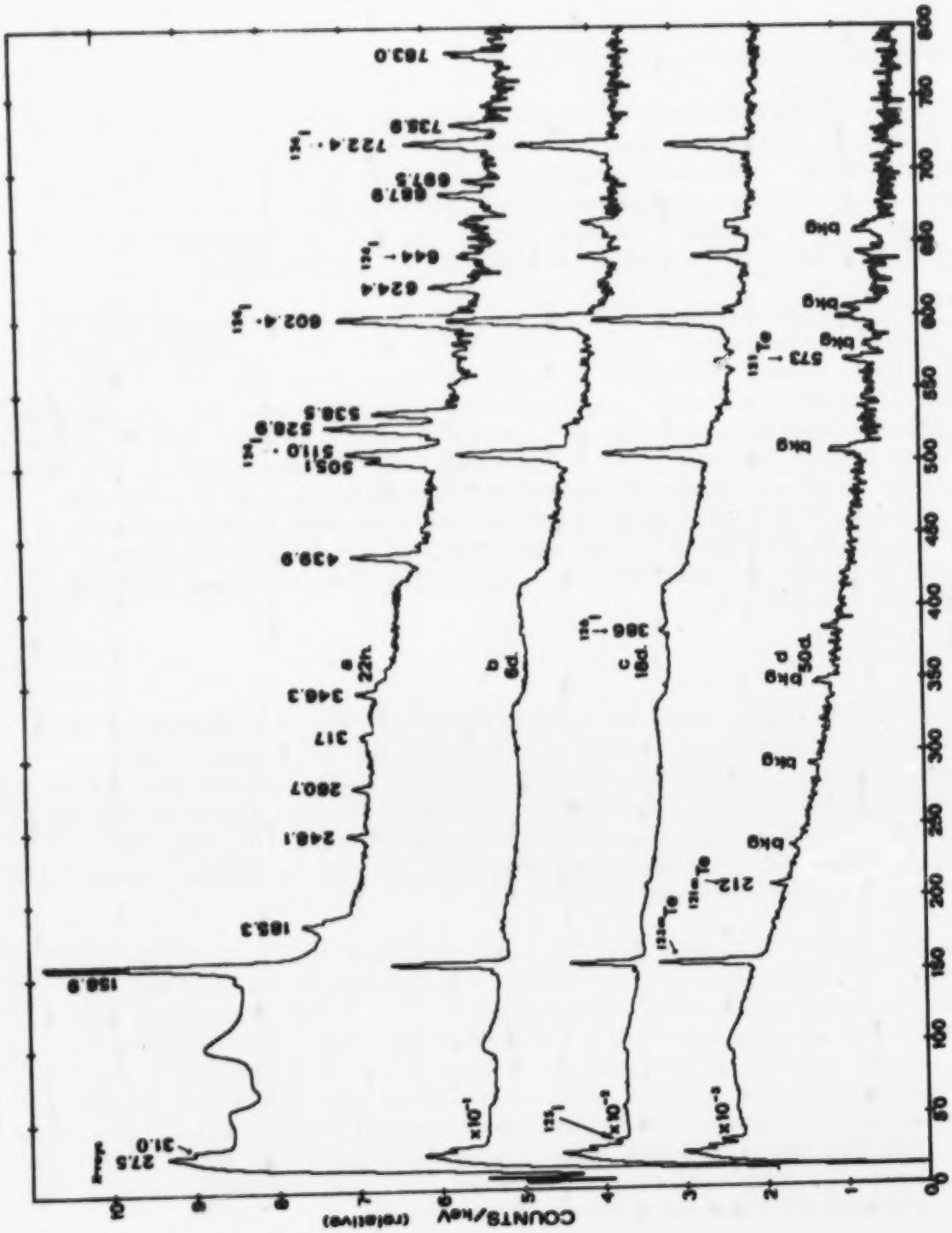

Figure 3. ^{123}I spectra from the $^{121}Sb(^{4}He,2n)$ ^{123}I reaction (I_a = 31 MeV)

The presence of 60 day ^{125}I and 12.8 day ^{126}I was confirmed in figure 3(c) by the identification of photopeaks at 35.4 and 388 keV, respectively. The ^{125}I and ^{126}I contaminations which resulted from $^{123}Sb(^{4}He,2n)\ ^{125}I$ and $^{123}Sb(^{4}He,n)\ ^{126}I$ reactions on the 1.6 percent ^{123}Sb impurity in the target were 1.5×10^{-2} and 2.6×10^{-3} percent.

In figure 3(d) the presence of 118 d ^{123m}Te, 154 d ^{121m}Te, and 17 d ^{121}Te was identified by photopeaks at 158.9 and 212 and 573 keV. The ^{123m}Te was produced from the $^{121}Sb(^{4}He,pn)\ ^{123m}Te$ reaction. The ^{123m}Te was found mainly in the target residue, but some was also carried over into the distillate.

The ^{121m}Te and ^{121}Te resulted either directly from $^{121}Sb(^{4}He,p3n)\ ^{121m}Te$ and ^{121}Te reactions or indirectly from the decay of ^{121}I produced from the $^{121}Sb(^{4}He,4n)\ ^{121}I$ reaction.

The results of bombarding thin (~15 mg/.7 cm^2) and thick (~50 mg/.7 cm^2) targets of 98.4 percent enriched ^{121}Sb with alpha particles ranging in energy from 25.2 - 36.5 MeV are listed in tables 4 and 5.

Tabulated are the bombardment energy, the weight of target per 0.7 cm^2, the ^{123}I yield (mCi/μA-h) and the percentages of ^{124}I, ^{125}I, and ^{126}I contaminants. All yields are corrected to the end of the bombardment. The energies listed are "average" energies, i.e., the average of the incident and exit energy of the beam traversing the target. The uncertainty is the difference between the average and the extreme values. The superficial density of the targets (mg/cm^2) can be calculated by dividing its weight by the target area, 0.7 cm. All bombardments were made with beam currents of 4 ± 2μA for 3-5 hours. The absolute ^{123}I yields from bombardment numbers 23, 30, and 33 are not listed because the current integrator was not operating during those runs. However, the percentages of ^{124}I, ^{125}I, and ^{126}I contaminants are listed because these relative values are independent of the total charge on the target.

Table 4. Radioiodine yields from the $^{121}Sb(^{4}He,2n)\ ^{123}I$ reaction on thin (~15 mg/0.7 cm^2) targets of ^{121}Sb

Run no.	Energy (MeV)	Target weight (mg/.7 cm^2)	^{123}I (mCi/μA-h)	^{124}I (percent)	^{125}I (percent x10^{-2})	^{126}I (percent x10^{-3})
4	28.4 ± 1.2	15.0	0.14	0.54	1.4	2.3
9	33.3 ± 1.1	15.8	0.23	0.62	1.5	1.9
5	33.4 ± 1.0	15.0	0.18	0.53	1.3	2.9
7	33.5 ± 0.9	13.4	0.16	0.54	1.4	2.3
10	36.5 ± 1.0	15.0	0.08	1.35	1.4	2.2

6.1.3 Discussion

The ^{123}I yields listed in table 4 in the thin ^{121}Sb targets appear to be similar in the bombardments made with 28.4 - 33.5 MeV alpha particles. The ^{123}I yield at this energy normalized to target weights of 15 mg was 0.14 - 0.22 mCi/μA-h. This energy range is between 25 MeV where the cross section for the $^{121}Sb(^{4}He,2n)$ reaction should be maximum and 34 MeV where that for the $^{121}Sb(^{4}He,3n)$ reaction should be maximum. Energies higher than 25 MeV were used to lessen the probability of the $^{121}Sb(^{4}He,n)$ ^{124}I reaction. This was advantageous since the $^{121}Sb(^{4}He,3n)$ reaction produced 3.5 m ^{122}I.

The ^{123}I yield, from the bombardment at higher energy (36.5 MeV) is lower.

The lowest ^{124}I contamination was 0.54 - 0.62 percent at bombardment energies from 28.4 to 33.5 MeV on 15 mg targets. The ^{124}I contamination increased from 0.6 to 1.3 percent when the bombardment energy was increased from 33.4 to 36.5 MeV.

This relative increase at higher energy resulted from a decrease in the probability of the $^{121}Sb(^{4}He,2n)$ ^{123}I reaction and an increase in the probability of the $^{123}Sb(^{4}He,3n)$ ^{124}I reaction.

Table 5. Radioiodine yields from the $^{121}Sb(^{4}He,2n)$ ^{123}I reaction on thick (~50 mg/0.7 cm^2) targets of ^{121}Sb

Run no.	Energy (MeV)	Target weight (mg/.7 cm^2)	^{123}I (mCi/μA-h)	^{124}I (percent)	^{125}I (percent x10^{-2})	^{126}I (percent x10^{-3})
2	25.2 ± 4.0	50.0	0.69	0.89	1.4	2.3
18	31.0 ± 3.4	49.6	0.48	1.04	1.6	3.0
20	31.0 ± 3.4	49.8	0.47	0.70	1.5	2.5
23	31.0 ± 3.4	50.0	(a)	0.68	1.4	--
30	31.0 ± 3.4	50.8	(a)	0.70	--	--
32	31.0 ± 3.4	49.9	0.80	0.65	1.2	2.0
33	31.0 ± 3.4	50.6	(a)	0.69	1.3	2.1
50	31.0 + 3.4	50.3	0.58	0.74	1.4	2.2
3	31.1 ± 3.3	46.5	0.93	0.84	1.5	3.0
41	31.5 ± 3.4	50.0	0.55	0.76	1.3	2.7
42	31.5 ± 3.4	50.2	0.38	0.87	1.8	2.9
46	31.5 ± 3.4	50.8	0.50	0.86	1.6	2.1
48	31.5 ± 3.4	50.8	0.75	1.04	1.4	3.2

[a]Current integrator in error.

The ^{123}I yields from 50 mg/.7 cm^2 ^{121}Sb targets listed in table 5 were 0.38 - 0.93 mCi/μA-h at the alpha particle bombardment energy of 31 ± 3 MeV. The average yield (0.012 mCi/μA-h-mg) for 50 mg targets is the same as that for 15 mg targets.

The ^{124}I contamination listed in table 5 was 0.65 - 1.06 percent with an average yield of 0.79 ± 0.13 percent. This average ^{124}I contamination was approximately 30 percent higher than that obtained with the 15 mg targets. The increase in the ^{124}I yield resulted from the increased probability of the $^{121}Sb(^4He,n)$ ^{124}I reaction occurring as the alpha particle energy was attenuated in traversing the thicker targets. The approximately linear relationship of ^{123}I yield vs. the weight of the ^{121}Sb targets from 15 - 50 mg, however, indicated that penetration through the 50 mg/.7 cm^2 target thickness did not attenuate the incident alpha particle energy (34 MeV) below that which caused the desired $^{121}Sb(^4He,2n)$ ^{123}I reaction.

Because of the linear increase of ^{123}I with target thickness, the ^{123}I yield per μA-h could probably have been increased further by using ^{121}Sb targets greater than 50 mg. Unfortunately, the ^{124}I contamination was increased in the thicker targets. Because minimum ^{124}I contamination is advantageous if the ^{123}I is to be used for low dose radiopharmaceutical use, bombardment of targets heavier than 50 mg was not attempted.

The average ^{125}I contamination listed in table 5 from 50 mg ^{121}Sb targets irradiated with 31 MeV alpha particles was $(1.5 \pm 0.2) \times 10^{-2}$ percent, the average ^{126}I contamination was $(2.6 \pm 0.4) \times 10^{-3}$ percent.

6.2 $^{121}Sb(^3He,n)$ ^{123}I

6.2.1 Experimental Considerations

The coulomb barrier for this reaction, 15 MeV, is 10 MeV higher than its E_{BP}. However, although this reaction has a low theoretical cross section, it was attempted for the following reasons:

a. Extremely high purity ^{121}Sb is available.

b. The low energy, 15 MeV, ^3He particles can be obtained from the low cost cyclotrons available to many nuclear medical institutions.

c. Theoretically, no ^{124}I contamination should be produced from the $(^3He,\gamma)$ reactions which have low probability at 5-15 MeV incident ^3He energies.

The bombardments were done at the Ohio State University cyclotron which accelerates ^3He particles to 18 MeV. The beam current was 2-4 μA. The ^{121}Sb target powder was bombarded in a beam area 0.22 cm^2.

6.2.2 Results

The results of bombarding 8-16 mg 98.4 percent ^{121}Sb targets with 4.5 - 14.0 MeV ^{3}He particles are listed in table 6. At 4.5 MeV the ^{123}I yeild was only 8.5×10^{-6} mCi/μA-h. The ^{124}I contamination was 2.7 percent. An attempt to raise the yield was made at 14 MeV resulting in about a 10-fold increase in ^{123}I. However, the ^{124}I increased to 4.3 percent and a great amount of ^{121}I was formed from the $^{121}Sb(^{3}He,3n)\ ^{121}I$ reaction.

6.2.3 Discussion

At 4.5 MeV, close to the E_{BP}, the coulomb barrier limited the amount of ^{123}I produced from the $(^{3}He,n)$ reaction to only 8.5×10^{-6} mCi/μA-h. This yield is too low for routine ^{123}I production. The ^{124}I was produced from $(^{3}He,2n)$ reactions on the 1.5 percent ^{123}Sb impurity in the target.

At 14.0 MeV, close to the coulomb barrier, the cross sections for the $(^{3}He,2n)$ and $(^{3}He,3n)$ reactions have increased more than that of the desired $(^{3}He,n)$ reaction as evidenced by the 20-fold increase in ^{124}I and the production of great amounts of ^{121}I (44 times more than the ^{123}I yield). Undoubtedly, a large amount of 3.5 m ^{122}I was also produced from the $^{121}Sb(^{3}He,2n)$ reaction, but had already decayed by the time of analysis.

It is obvious that this reaction cannot be used for production of radiopharmaceutical ^{123}I.

Table 6. Radioiodine yields from the $^{121}Sb(^{3}He,n)\ ^{123}I$ reaction on 98.4 percent ^{121}Sb

Run no.	Energy (MeV)	Target weight (mg/.2 cm^2)	^{123}I (mCi/μA-h)	^{124}I (percent)	^{121}I (mCi/μA-h)
16	4.5 ± 4.5	15.9	8.5×10^{-6}	2.7	--
13	14.0 ± 2.5	7.7	1.1×10^{-4}	4.3	4.7×10^{-3}

6.3 $^{122}Te(d,n)\ ^{123}I$

6.3.1 Experimental Considerations

The coulomb barrier for the $^{122}Te(d,n)\ ^{123}I$ reactions, 8 MeV, is close to the E_{BP}, 7 MeV. The reaction was attempted at 6.4 - 9.0 MeV.

In favor of this reaction, the radioiodine contamination is limited only by (d,n) and (d,2n) reactions on tellurium impurities in the target and not by competing reactions on ^{122}Te. The $^{122}Te(d,\gamma)\ ^{124}I$ reaction is highly improbable at these bombardment energies; the $^{122}Te(d,2n)\ ^{122}I$ reaction produces the 3.5 m contaminant.

Inquiry to two suppliers of ^{122}Te showed that it was economically impractical to reduce the tellurium impurities to less than 5 percent.

6.3.2 Results

Two typical Ge(Li) spectra of a radioiodine fraction distilled from a 95.4 percent enriched ^{122}Te target are shown in figure 4. Figure 4(a and b) were obtained 7.8 hours and 9 days, respectively, after the end of the bombardment. The deuteron energy of this bombardment was 6.8 $\pm$ 0.5 MeV. No major activity was found above 800 keV.

Identified in figure 4(a) are the 27.5 and 31.0 keV x-rays and the 158.9, 248.1, 280.7, 346.3, 439.9, 505.1, 528.9, 624.4, and 783.0 keV gamma-ray photopeaks of ^{123}I. The 185.3 and 317 keV photopeaks are sums of abundant photons as described above for figure 3(a). Also identified are the gamma-ray photopeaks of ^{124}I at 602.4 and 722.4 keV, and of ^{130}I at 417.9, 536.1, 668.2, and 739.2 keV. The ^{124}I and ^{130}I contaminations which resulted principally from $^{123}Te(d,n)\ ^{124}I$ and $^{130}Te(d,2n)\ ^{130}I$ reactions on the tellurium impurities in the target were 0.083 and 0.92 percent, respectively, at the end of this particular bombardment.

Identified in figure 4(b) are the gamma-ray photopeaks of ^{125}I at 35.4 keV, of ^{126}I at 388.6 and 666.4 keV and of ^{131}I at 80.2, 284.3, 364.5, and 637.0 keV. The ^{125}I, ^{126}I, and ^{131}I contaminants which result mainly from $^{124}Te(d,n)\ ^{125}I$, $^{125}Te(d,n)\ ^{126}I$, and $^{130}Te(d,n)\ ^{131}I$ reactions were 0.041, 0.039, and 0.07 percent, respectively, at the end of this particular bombardment. Also identified in figure 4(b) is the 511 keV annihilation photopeak caused primarily by ^{124}I. The photopeak at 158.9 keV was caused by residual ^{123}I and 118 day ^{123m}Te, the latter persisting long after the 13.3 hour ^{123}I has disappeared. The ^{123m}Te was produced by decay of ^{123}I or was carried over during the distillation despite the presence of hold-back carrier.

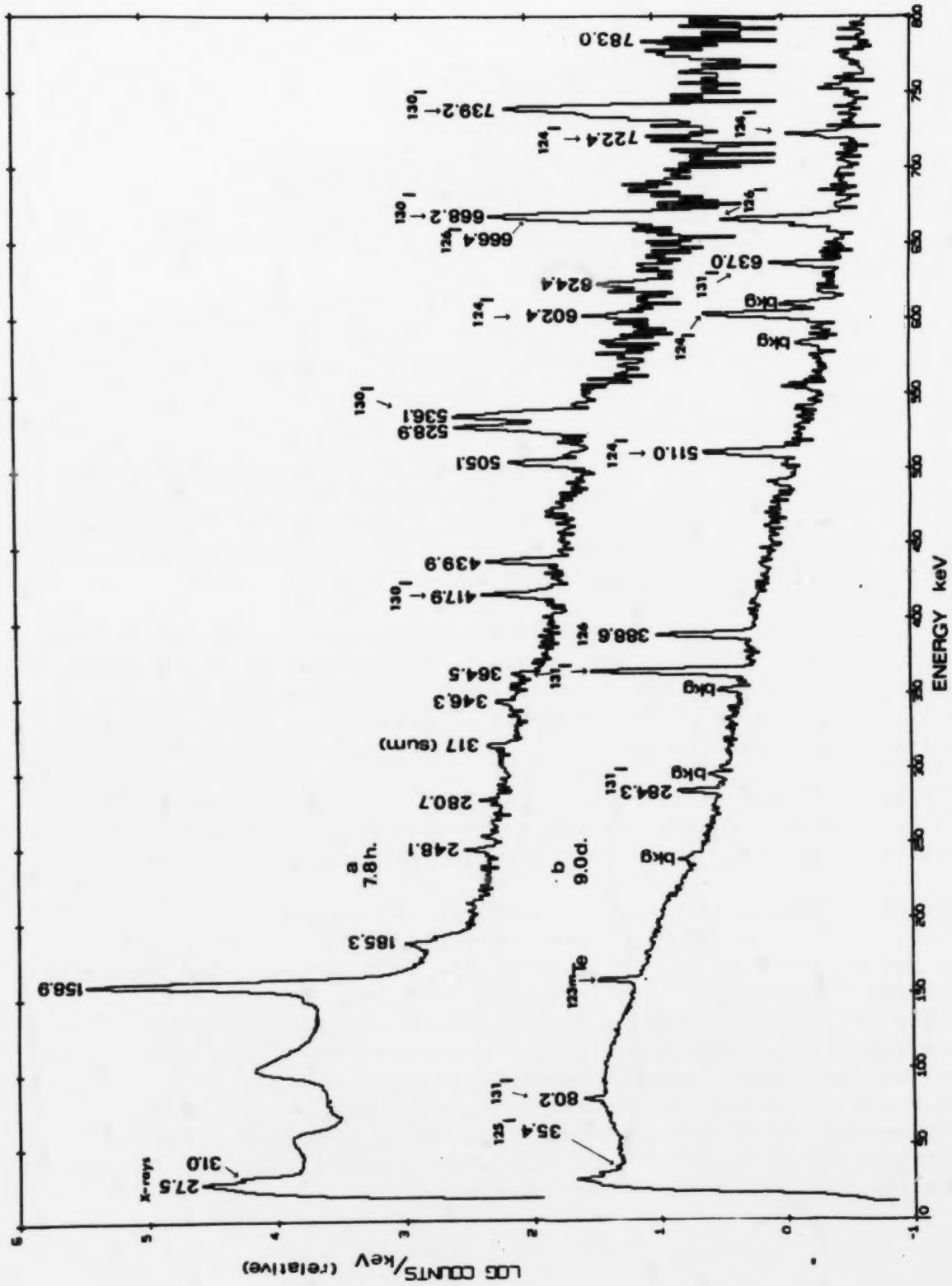

Figure 4. ^{123}I spectra from the $^{122}Te(d,n)$ ^{123}I reaction (E_d = 6.8 MeV)

The radioiodine yields from 6.4 - 9.0 MeV deuteron bombardments of 95.4 percent enriched ^{122}Te are listed in table 7. Also listed for each bombardment are the "average" energy of the deuteron beam, the weight of the target per 0.7 cm^2, and the percentages of ^{124}I, ^{125}I, ^{126}I, ^{130}I, and ^{131}I contaminants. The bombardments were made with beam currents ranging from 4-7 μA for 1-6 hours. Bombardment no. 28 was actually done on 80.4 percent enriched ^{122}Te. The results shown in table 7 for this run were corrected to those which would be obtained from bombardment of 95.4 percent enriched ^{122}Te. The corrections were made assuming the ^{123}I, ^{124}I, ^{125}I, ^{126}I, ^{130}I, and ^{131}I were produced from $^{122}Te(d,n)\ ^{123}I$, $^{123}Te(d,n)\ ^{124}I$, $^{124}Te(d,n)\ ^{125}I$, $^{125}Te(d,n)\ ^{126}I$, $^{130}Te(d,n)\ ^{131}I$, and $^{130}Te(d,2n)\ ^{130}I$ reactions, respectively.

Table 7. Radioiodine yields from the $^{122}Te(d,n)\ ^{123}I$ reaction on 95.4 percent ^{122}Te

Run no.	Energy (MeV)	Target weight (mg/.7 cm^2)	^{123}I (mCi/μA-h x10^{-2})	^{124}I (percent x10^{-2})	^{125}I (percent x10^{-2})	^{126}I (percent x10^{-2})	^{130}I (percent)	^{131}I (percent)
39	6.4 ± 0.3	12.5	0.19	7.2	1.6	4.1	0.97	0.15
34	6.8 ± 0.5	17.3	1.5	8.3	2.1	3.9	0.92	0.07
38	6.8 ± 0.5	16.3	10.1	8.5	1.3	4.4	0.86	0.09
29	6.9 ± 0.4	15.8	6.2	7.2	2.1	3.4	0.82	0.08
[a]28	7.3 ± 1.0	31.5	13.2	19.2	1.2	6.6	0.96	0.07
31	9.0 ± 0.3	12.6	8.1	24.0	1.3	9.6	2.5	0.09

[a]Bombardment was performed using 80.4 percent ^{122}Te; yields shown here have been corrected to 95.4 percent ^{122}Te.

6.3.3 Discussion

The ^{123}I yields listed in table 7 from 6.8 - 7.3 MeV deuteron bombardment of 15.8 - 31.5 mg ^{122}Te targets, i.e., from bombardment nos. 28, 29, 34, and 38, varied considerably from (1.5 to 13.2) x 10^{-2} mCi/μA-h. The large variation in yield from the $^{122}Te(d,n)\ ^{123}I$ reaction is probably related to the fact that the calculated coulomb barrier for the reaction is approximately 8 MeV. Assuming that the cross section of the $^{122}Te(d,n)\ ^{123}I$ reaction is smooth function of the excitation energy, it was estimated that the cross section should be approximately maximum at the theoretical threshold for the $^{122}Te(d,2n)\ ^{122}I$ reaction, i.e., about 7 MeV. However, because the coulomb barrier opposes the approach of the deuteron to the ^{122}Te nucleus at this energy, the cross section for the reaction will probably actually be a very steep function of the energy. This

supposition is borne out by the 0.19 x 10^{-2} mCi/μA-h ^{123}I yield obtained at 6.4 MeV. This yield is 10-100 times less than those obtained at 1 MeV higher bombardment energies. The large variations in ^{123}I yields under seemingly identical conditions are probably due to the unavoidable variations in the thicknesses of the powder targets which in some instances reduce the excitation energy significantly below the coulomb barrier.

The ^{123}I yield at 9.0 MeV deuteron-bombardment energy, about 1 MeV higher than the calculated coulomb barrier, was approximately the same as those obtained at 6.8 - 7.3 MeV. The yields of ^{124}I, ^{126}I, and ^{130}I, however, have increased 2-3 fold at this higher excitation energy because of the increased cross sections of the ^{124}Te(d,2n) ^{124}I, ^{126}Te(d,2n) ^{126}I, and ^{130}Te(d,2n) ^{130}I reactions, respectively. The results show that the highest and purest yields of ^{123}I were obtained with 7 MeV deuteron bombardment of 15 mg 95.4 percent ^{122}Te targets.

6.4 ^{122}Te(^{4}He,3n) ^{123}Xe and ^{122}Te(^{4}He,p2n) ^{123}I

6.4.1 Experimental Considerations

The E_{BP} for these reactions, 35 MeV, is well above their coulomb barriers at 15 MeV. These reactions were studied together by entrapping the ^{123}Xe gas in the sealed standard target (see section 3.3) along with the directly produced ^{123}I. The sealed target was opened after the ^{123}Xe had decayed to ^{123}I.

6.4.2 Results

The results of bombarding 95.4 percent ^{122}Te with 40 MeV alpha particles are listed in table 8. The ^{123}I yield from this reaction was .16 - .25 mCi/μA-h, the yield increasing as the target thickness increased. The ^{123}I yields were corrected to 2-4 hours after bombardment when ingrowth from ^{123}Xe was maximum. The ^{124}I, ^{125}I, and ^{126}I contaminants were 1.1 - 1.6, 0.03 - 0.06, and 0.01 percent, respectively. With the generator technique, except for small amounts of ^{125}I (0.2 percent), the longer lived radioiodine contaminants are eliminated.

Table 8. Radioiodine yields from the ^{122}Te(^{4}He,3n) ^{123}Xe$\longrightarrow$ ^{123}I and ^{122}Te(^{4}He,p2n) ^{123}I reactions on 95.4 percent ^{122}Te

Run no.	Energy (MeV)	Target weight (mg/.7 cm^2)	^{123}I (mCi/μA-h)	^{124}I (percent)	^{125}I (percent)	^{126}I (percent)
2a	40.4 ± 0.7	11.3	0.16	1.1	0.03	0.006
6	40.0 ± 1.1	17.2	0.21	1.6	0.06	0.004
8	39.5 ± 1.6	25.0	0.25	1.5	0.03	0.009

6.4.3 Discussion

The ^{123}I yield from 40 MeV ^{4}He reactions on ^{122}Te is about 0.012 mCi/μA-h-mg of target at target thicknesses of 17-36 mg/cm^2. The incident alpha particles at 41 MeV will degrade in energy to 34 MeV in penetrating a 75 mg/cm^2 ^{122}Te target. If 35 MeV is the E_{BP} for these reactions, one can assume a linear increase of ^{123}I yield with targets at least up to 75 mg/cm^2 in thickness. Therefore, the projected ^{123}I yield from these reactions is about 0.65 mCi/μA-h.

6.5 $^{122}Te(^{3}He,2n)\ ^{123}Xe$ and $^{122}Te(^{3}He,pn)\ ^{123}I$

6.5.1 Experimental Considerations

The coulomb barriers for $^{122}Te(^{3}He,2n)\ ^{123}Xe$ and $^{122}Te(^{3}He,pn)\ ^{123}I$ reactions at 16 MeV are higher than the 14 and 13 MeV respective E_{BP}'s. Therefore, these reactions should be expected to have limited yields. However, the $^{122}Te(^{3}He,2n)\ ^{123}Xe$ reaction was attempted because it was within the energy limitations of the compact cyclotron allowing use of the generator technique with the smaller machine. The compact cyclotron cannot accelerate alpha particles to the 35-40 MeV necessary to cause the $^{122}Te(^{4}He,3n)\ ^{123}Xe$ reaction.

Two methods of studying these reactions were used. The target from the bombardments at ANL was allowed to decay several hours and the ^{123}I was chemically separated according to the procedure in appendix 1. The total ^{123}I yield from both reactions was corrected to the time of maximum ^{123}I ingrowth from ^{123}Xe.

The targets from the bombardments at SKL were counted without chemical treatment to individually assay the $^{122}Te(^{3}He,2n)\ ^{123}Xe$ and $^{122}Te(^{3}He,pn)\ ^{123}I$ reactions. The total ^{123}I yield was then calculated at the optimal ingrowth time from the individual yields.

6.5.2 Results

The ^{123}I yields from two sealed 80 percent ^{122}Te targets bombarded with 14.5 MeV ^{3}He particles at ANL are listed in table 9. The ^{123}I yield from 9.8 - 13.8 mg ^{122}Te targets was (1.8 - 2.3) x 10^{-2} mCi/μA-h. The ^{124}I contamination averages 0.7 percent.

Table 9. Radioiodine yields from the $^{122}Te(^{3}He,2n)\ ^{123}Xe$ and $^{122}Te(^{3}He,pn)\ ^{123}I$ reactions on 80.4 percent ^{122}Te

Run no.	Energy (MeV)	Target weight (mg/.7 cm^2)	^{123}I (mCi/μA-h x10^{-2})	^{124}I (percent)
36	14.7 ± .9	9.8	2.3	0.63
37	14.2 ± 1.4	13.8	1.8	0.83

The ^{123}I, ^{123}Xe, and total available ^{123}I yields obtained from 14.9 and 20.7 MeV ^{3}He bombardment of 73.3 percent ^{122}Te are shown in table 10. The ^{123}Xe and ^{123}I yields after the end of the bombardment with 13.5 MeV ^{3}He particles were 0.18×10^{-3} and 0.06×10^{-3} mCi/μA-h, respectively. At 17.9 MeV, the ^{123}Xe and ^{123}I yields were 13.7×10^{-3} and 6.3×10^{-3} mCi/μA-h, respectively.

6.5.3 Discussion

The total ^{123}I yields listed in table 10 are the calculated maximal amounts of ^{123}I available from the target at the optimum ingrowth time. These values can be compared to those listed in table 9 where the total ^{123}I yield was measured directly. In making this comparison, the following inequalities must be considered:

a. Because the ^{122}Te enrichment was less for the bombardments in table 10, these results should be increased about 10 percent.

b. Because of the lack of beam directing capabilities at SKL, only 60-70 percent of the entire target was irradiated.

c. Target weights were different in the bombardments.

The total ^{123}I yield at 13.5 MeV is 100-fold less than that at 14.7 MeV. This great decrease in ^{123}I yield at slightly lower energy is due to factors related to the 16 MeV coulomb barrier for this reaction (see section 6.3.3). The ^{123}I yields for the three runs made with 14.2 - 17.9 MeV ^{3}He particles are comparable at 0.02 mCi/μA-h-mg of target.

The ^{123}Xe yield/mg of target at 17.8 MeV is 100-fold higher than at 13.5 MeV. However, the ^{123}I yield is 150-fold higher. This can be explained by the increasing probability of charged particle emission from the compound nucleus at the higher energy. To compare the ^{123}I yields from the direct and indirect sources, the ^{123}Xe yields must be divided by 6.3, i.e., $[t_{1/2}(^{123}I)]/t_{1/2}(^{123}Xe)$. The ^{123}I yield available from the $^{122}Te(^{3}He,pn)\ ^{123}I$ reaction is about 2 to 3 times greater than that available indirectly from the $^{122}Te(^{3}He,2n)\ ^{123}Xe$ reaction.

Table 10. ^{123}Xe, ^{123}I and total available ^{123}I from the $^{122}Te(^{3}He,2n)\ ^{123}Xe$ and $^{122}Te(^{3}He,pn)\ ^{123}I$ reaction on 73.3 percent ^{122}Te

Run no.	Energy (MeV)	Target weight (mg/.7 cm^2)	Yield (mCi/μA-h x 10^{-3})		
			^{123}Xe	^{123}I	Total ^{123}I
43 - 2B	13.5 ± 0.8	7.8	0.18	0.06	0.07
43 - 2A	17.9 ± 0.6	5.4	13.7	6.3	6.7

6.6 $^{123}Te(^{3}He,3n)\ ^{123}Xe$ and $^{123}Te(^{3}He,p2n)\ ^{123}I$

6.6.1 Experimental Considerations

The E_{BP}'s of the $^{123}Te(^{3}He,3n)\ ^{123}Xe$ and $^{123}Te(^{3}He,p2n)\ ^{123}I$ reactions at 21 and 20 MeV, respectively, are well above their 16 MeV coulomb barriers.

In favor of this reaction over other ^{123}Xe producing reactions, i.e., $^{122}Te(^{4}He,3n)\ ^{123}Xe$ and $^{122}Te(^{3}He,2n)\ ^{123}Xe$ are the following:

a. The 25 MeV ^{3}He particle necessary for this reaction is lower in energy than the 42 MeV ^{4}He particle necessary for the $^{122}Te(^{4}He,3n)\ ^{123}Xe$ reaction. The former is within the energy limitations of some compact cyclotrons whereas the latter is not.

b. Because ^{3}He induced reactions on ^{123}Te and ^{4}He induced reactions on ^{122}Te form the same compound nucleus, ^{126}Xe, identical yields should be obtained.

c. No coulomb barrier interference exists as there does with the $^{122}Te(^{3}He,2n)\ ^{123}Xe$ and the $^{122}Te(^{3}He,pn)\ ^{123}I$ reactions.

d. Because the desired ^{3}He induced reactions on ^{123}Te require higher-energy ^{3}He particles than do those on ^{122}Te, thicker targets can be used.

The disadvantages of those reactions using ^{123}Te over those using ^{122}Te are the following:

a. The cost of ^{123}Te is more than triple that of ^{122}Te.

b. The isotopic enrichments of ^{123}Te are available only up to 80 percent, while those of ^{122}Te are up to 95 percent.

6.6.2 Results

The results of bombarding 6 mg targets of ^{123}Te with 16.8 and 21.4 MeV ^{3}He particles are listed in table 11. The ^{123}Xe yield at the end of the bombardment at 16.8 MeV was 1.6×10^{-3} mCi/μA-h; at 21.4 MeV was 7.7×10^{-3} mCi/μA-h. The directly produced ^{123}I yield at the end of the bombardment at lower energy was comparable to that at higher energy with yields of 0.56×10^{-3} and 0.78×10^{-3} mCi/μA-h, respectively.

Table 11. ^{123}Xe, ^{123}I and total available ^{123}I from the $^{123}Te(^{3}He, 3n)\ ^{123}Xe$ and $^{123}Te(^{3}He,p2n)\ ^{123}I$ reactions on 76.5 percent ^{123}Te

Run no.	Energy (MeV)	Target weight (mg/.7 cm^2)	Yield (mCi/μA-h x 10^{-3})		
			^{123}Xe	^{123}I	Total ^{123}I
1B	16.8 ± 0.5	5.4	1.6	0.56	0.60
1A	21.4 ± 0.4	6.0	7.7	0.78	1.44

6.6.3 Discussion

The maximum available ^{123}I yields calculated to the time of optimum ingrowth can be compared to the total ^{123}I yields from the $^{122}Te(^{4}He,3n)\ ^{123}Xe$ and $^{122}Te(^{4}He,p2n)\ ^{123}I$ reactions which were measured directly (see table 8). The total ^{123}I yield from the ^{3}He induced reactions on ^{123}Te is only 2 percent of that from the ^{4}He induced reactions on ^{122}Te. However, the authors feel that the ^{123}I yields from ^{3}He induced reactions on ^{123}Te could be increased by using particles higher in energy than the 22 MeV ^{3}He which were available for this work. Further study of these reactions will be done at 28 MeV. Assuming that the compound nucleus, ^{126}Xe, is formed both with the ^{3}He induced reactions on ^{123}Te and the ^{4}He induced reactions on ^{122}Te, then comparable ^{123}Xe and ^{123}I yields should be expected.

An interesting observation is that the ^{123}Xe yield increased 5 fold at the higher energy whereas, the ^{123}I yield increased only 39 percent. If this trend continues at still higher energies, then most of the available ^{123}I would be produced indirectly from ^{123}Xe decay.

6.7 USE OF GAS-FLOW TARGET ASSEMBLY

6.7.1 Experimental Considerations

The continuous gas-flow target as described in section 3.4 was used with the $^{122}Te(^{4}He,3n)\ ^{123}Xe$ reaction. The continuous flow principle can also be used with the other ^{123}Xe producing reactions described in sections 6.5 and 6.6.

Isolation of xenon from ^{3}He and ^{4}He bombardment of tellurium is feasible for pure ^{123}I production for the following reasons:

a. Xenon can be physically separated from other elements.

b. The ^{124}Xe, ^{126}Xe, ^{128}Xe, ^{129}Xe, ^{130}Xe, ^{131}Xe, ^{132}Xe, and ^{134}Xe isotopes are stable.

c. The 36.4 d ^{127}Xe and 5.3 d ^{133}Xe isotopes decay to stable daughters.

d. Xenon isotopes with atomic mass less than 122 decay with short half-lives ≤41 minutes to short half-life radioiodine daughters ≤2 hours.

e. The 19 h ^{122}Xe decays to 3.5 m ^{122}I.

f. The major radioiodine contaminant is 60 d ^{125}I which results from decay of 17 h ^{125}Xe. Separation of ^{123}I from ^{123}Xe after only 2-5 hours decay prevents the ingrowth of most of the available ^{125}I. The ^{125}I contaminant decays by electron capture and emits 35 keV gamma-rays.

Furthermore, the gas-flow method is advantageous over the conventional sealed target-chemical separation method for these additional reasons:

a. The ^{123}Xe decays to monatomic ^{123}I which is chemically very reactive. This presents opportunities for the direct labeling of organ specific compounds.

b. The time necessary for ^{123}I ingrowth can be used to transport the isotope to the nuclear medical facility.

c. The target material does not have to be recovered.

d. The preparation of radiopharmaceutical ^{123}I by simply leaching the trap eliminates the standard iodine distillation.

e. Eliminates handling of non-^{123}I radioactivity normally encountered with bombardment of standard targets.

6.7.2 Results

The results of the continuous flow generator using the $^{122}Te(^{4}He,3n)\ ^{123}Xe$ reaction are shown in table 12. Runs no. 12 and 21 were done on 95.4 percent ^{122}Te, whereas no. 26 was done on natural tellurium which contains 2.5 percent ^{122}Te.

The ^{123}I yield from 100-127 mg of 95.4 percent ^{122}Te spread over an area of 6 cm^2 averaged about 0.045 mCi/μA-h. The yield from 10 g natural tellurium in the gas-flow generator was 0.003 mCi/μA-h.

Using the cyclic method (see section 3.4) in run 25 on 140 mg of natural tellurium in a standard target holder yielded 0.005 mCi/μA-h.

Table 12. Radioiodine yields of the $^{122}Te(^{4}He,3n)\ ^{123}Xe$ reaction using the continuous flow generator

Run no.	Target enrichment (percent)	Energy (MeV)	Target weight (mg/6 cm^2)	^{123}I (mCi/μA-h)	^{124}I (percent)
12	95.4	40.6 ± 0.5	127	0.047	0.27
21	95.4	40.7 ± 0.4	100	0.042	[a]0.04
26	2.5	42	10,000	0.003	--
[b]25	2.5	42	[c]140	0.005	[a]0.05

[a]Ten turn CO_2 trap
[b]Cyclic method
[c]Target weight in mg/.7 cm^2

6.7.3 Discussion

In run 12 with a helium flow rate of 10 1/min and a copper U-trap to remove contaminants, the ^{124}I was 0.27 percent. However, with the 10-turn copper coil used in runs 21 and 25, the ^{124}I was reduced to 0.05 percent. Other runs using the single pass method reduced the ^{124}I to 0.005 percent. Unfortunately, due to the extremely slow helium flow, 25 ml/min, there was insufficient target cooling. This led to transport of tellurium from the target and poor ^{123}I yields. Better target assemblies using more efficient water cooling of the powder could probably be devised.

Allowing the xenon trap to decay only 3-4 hours optimized the ^{123}I yield and kept ^{125}I ingrowth low. The ^{125}I contamination was limited to less than 0.2 percent. A spectrum of high purity ^{123}I is shown in figure 5.

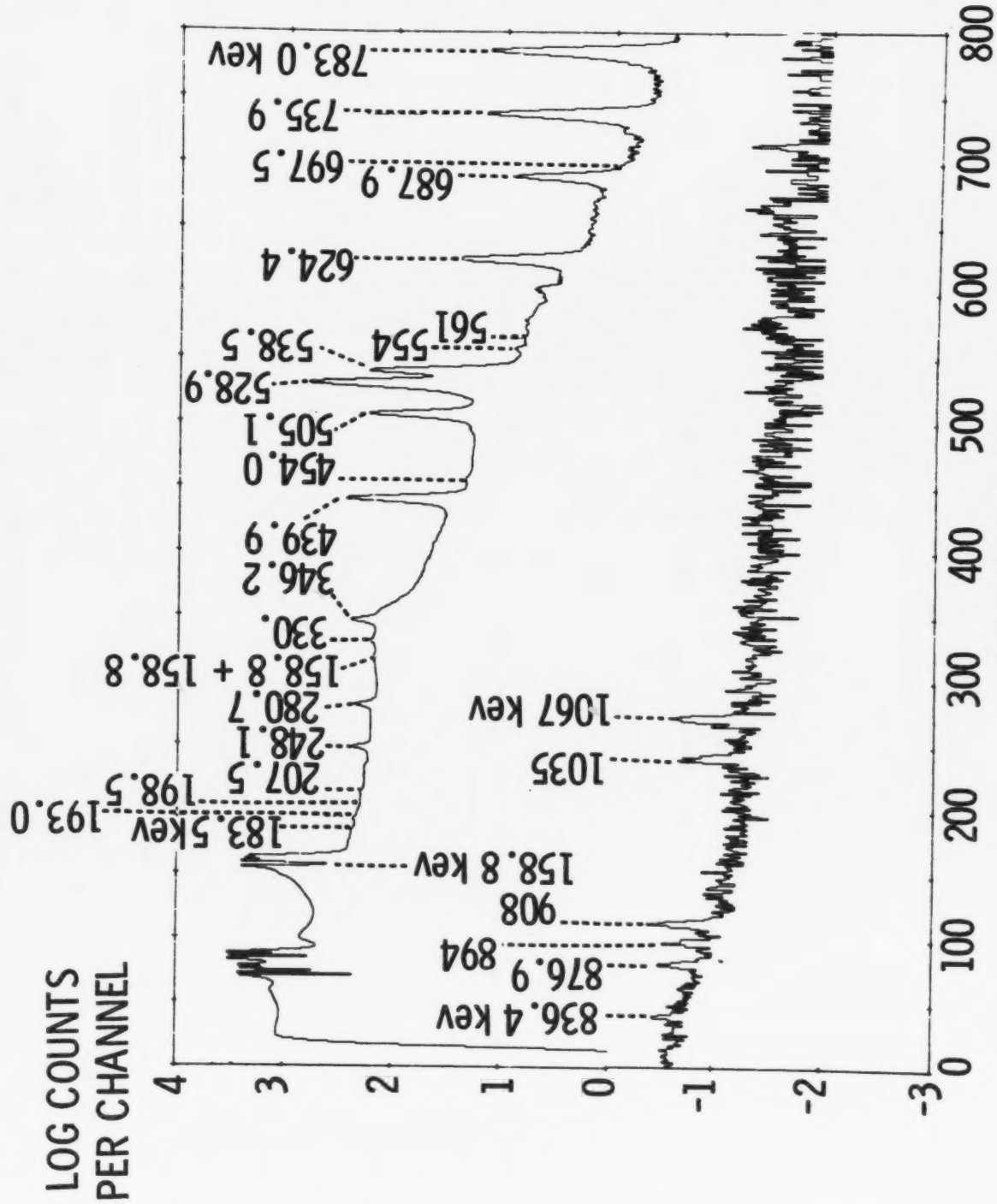

Figure 5. Ge(Li) spectrum of high purity ^{123}I

7. SUMMARY AND CONCLUSIONS

7.1 DIRECT METHODS

The $^{121}Sb(^{4}He,2n)\ ^{123}I$ reaction will provide satisfactory yields, 0.60 mCi/μA-h, for most needs. However, the 0.8 percent ^{124}I contamination limits the product's usefulness in high resolution nuclear medical applications and the need for 34 MeV ^{4}He particles restricts the production to higher energy cyclotrons.

By bombarding 95.4 percent ^{122}Te with 7 MeV deuterons in the $^{122}Te(d,n)\ ^{123}I$ reaction, the high-energy long-lived radioiodine contaminants can be kept to 0.3 percent and the ^{130}I to 0.9 percent. This method is useful with the "compact" cyclotron, but the yield is limited to about 0.1 mCi/μA-h by range-energy considerations. The short range of 7 MeV deuterons in tellurium restricts target thickness to less than 25 mg/cm^2.

The $^{121}Sb(^{3}He,n)\ ^{123}I$ is not practical at any energy. However, the $^{121}Sb(^{3}He,3n)\ ^{121}I$ might prove suitable for production of ^{121}I with the "compact" cyclotron.

The usefulness of the $^{122}Te(^{4}He,p2n)\ ^{123}I$, $^{122}Te(^{3}He,pn)\ ^{123}I$, and $^{123}Te(^{3}He,p2n)\ ^{123}I$ reactions is doubtful since the ^{124}I contaminations are greater than 1 percent.

7.2 INDIRECT METHODS

The ^{123}Xe producing reactions were studied for use with the generator technique which eliminates the longer-lived high-energy radioiodine contaminants. Nuclear medicine demands the highest purity ^{123}I to both improve scanning resolution and lower radiation exposure.

The $^{122}Te(^{4}He,3n)\ ^{123}Xe$ produces the highest yield of pure ^{123}I, .2 mCi/μA-h. This is using 41 MeV ^{4}He particles on 75 mg/cm^2 95.4 percent ^{122}Te. No "compact" cyclotron will accelerate 41 MeV ^{4}He particles.

The $^{122}Te(^{3}He,2n)\ ^{123}Xe$ reaction requires less than 20 MeV ^{3}He particles, which are produced with the "compact" cyclotron. However, because of this reaction's coulomb barrier problem and range-energy considerations, the yield is too low for use with the generator.

The $^{123}Te(^{3}He,3n)\ ^{123}Xe$ reaction has no coulomb barrier problem and needs a 28 MeV ^{3}He beam which is available from some "compact" cyclotrons. The yield from this reaction should be comparable to the $^{122}Te(^{4}He,3n)\ ^{123}I$, since the compound nucleus, ^{126}Xe, is the same. However, this could not be proven since the authors had only a 22 MeV beam available. Bombardments at higher energy could verify this.

The authors believe that the indirect methods of ^{123}I production are better than the direct methods because they eliminate chemical processing procedures while producing pure ^{123}I.

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APPENDIX

CHEMICAL PROCEDURES

1. Distillation of ^{123}I

The distillation apparatus used for the separation of ^{123}I is shown in figure 6. The target powder (10-50 mg) was scraped from the target holder and transferred with the aid of 0.5 ml iodide carrier solution (200µg I^-/ml) to a 300 ml Erlenmeyer flask. The standard Erlenmeyer flask was connected through ground glass joints to a 100 ml separatory funnel, a 12-inch water-cooled condenser and a 300 ml bubbling receiving vessel. The powder was dissolved in 12 ml hot concentrated H_2SO_4. The solution was cooled and diluted with 50 ml distilled water. The iodine then was distilled into the receiving vessel containing 25 ml 0.5 N NaOH. The yield of this distillation was determined by gamma counting the relative amounts of radioiodine both in the distillate and the residue. The distillation yield from fifteen ^{121}Sb targets was 91.2 $\pm$3.1 percent. High, but erratic, iodine yields (75 percent) were obtained when this same procedure was used without carrier.

2. Recovery of Target Material

When the availability and cost of the target materials were considered important factors, the enriched Sb and Te isotopes were recovered from composite residues.

a. Antimony

The acidic residue from the iodine distillation containing about 100 mg of ^{121}Sb was diluted with distilled water, neutralized with NH_4OH, and treated with 3 ml of hydrazine hydrate ($N_2H_4 \cdot H_2O$). The solution was adjusted in acidity to pH 1 with H_2SO_4 and saturated at room temperature with gaseous H_2S. The Sb_2S_3 precipitate was collected by centrifugation and washed with 1 N H_2SO_4. The sulfide was then dissolved in 10 ml of concentrated HCl and diluted with 10 ml of distilled water, and filtered through a glass-fiber filter. The clear solution was treated with chromous chloride solution to reduce the antimony to the metal, which was then collected by centrifugation, washed with ethyl alcohol and dried at 100° C. The chemical yield determined with natural antimony and ^{125}Sb tracer was 94.8 $\pm$1.5 percent.

b. Tellurium

The tellurium was recovered by essentially the same procedures reported by Hupf, Eldridge, and Beaver (*8*). The H_2SO_4 solution containing the ^{122}Te was adjusted to 1M by the addition of

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saturated $(NH_4)_2\ CO_3$ and then filtered. The solution was treated with 5 g NaBr and brought to a boil. The solution was then cooled. Three grams of $Na_2\ SO_3$ in divided portions were added slowly. The metallic ^{122}Te was collected by centrifugation, washed with water and dried at 110° C. The chemical yield of the procedure was 80 percent.

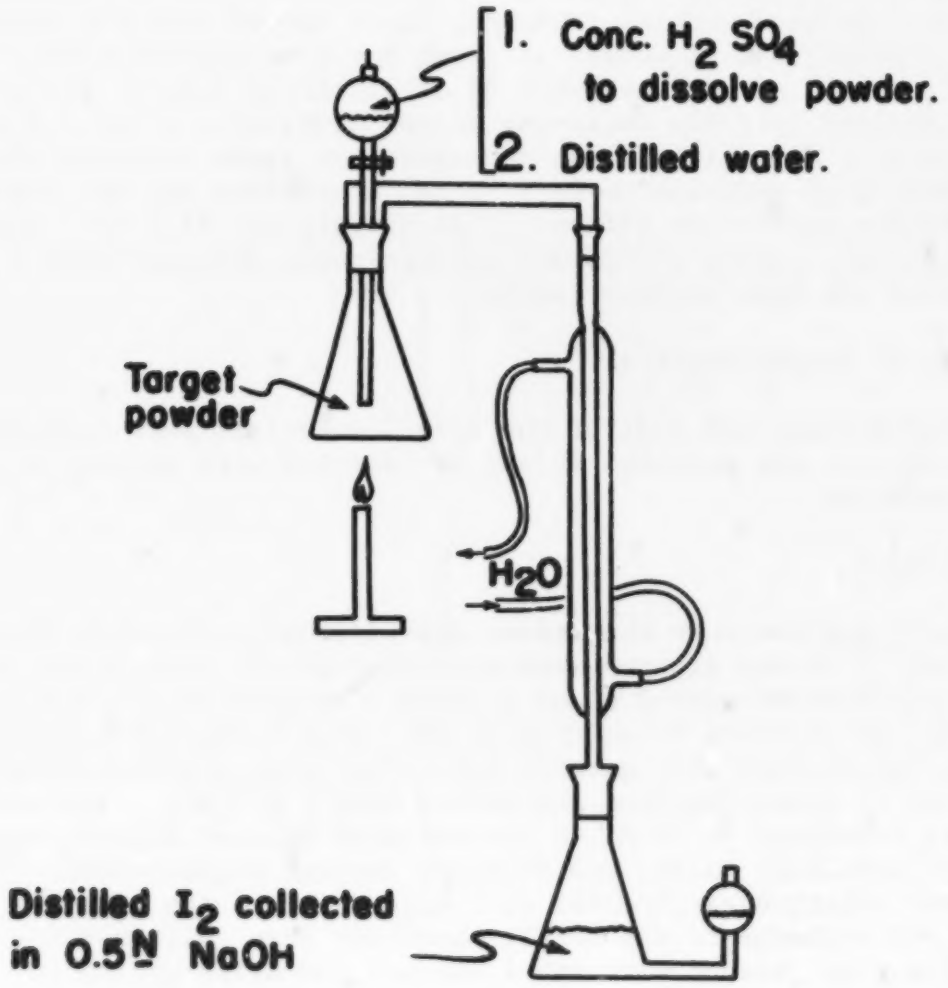

Figure 6. ^{123}I distillation apparatus

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V. J. SODD, K. L. SCHOLZ, J. W. BLUE, and H. N. WELLMAN:
Cyclotron Production of ^{123}I - an Evaluation of the Nuclear Reactions Which Produce This Isotope.
U.S. Department of Health, Education, and Welfare, Public Health Service Publication Number BRH/DMRE 70-4 (October 1970) 38 pp. (limited distribution).
ABSTRACT: Studied in this work were the $^{121}Sb(^{4}He,2n)$ ^{123}I, $^{121}Sb(^{3}He,n)$ ^{123}I, $^{122}Te(d,n)$ ^{123}I, $^{122}Te(^{4}He,p2n)$ ^{123}I, $^{122}Te(^{3}He,pn)$ ^{123}I and $^{123}Te(^{3}He,p2n)$ ^{123}I reactions that produce ^{123}I directly and the $^{122}Te(^{4}He,3n)$ ^{123}Xe, $^{122}Te(^{3}He,2n)$ ^{123}Xe and the $^{123}Te(^{3}He,3n)$ ^{123}Xe reactions that produce ^{123}I

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indirectly through the positron decay of 2.1 hour ^{123}Xe. The authors recommend the use of the gas-flow ^{123}I cyclotron target assembly for the production of ^{123}I with radiochemical purity greater than 99.995%.

KEYWORDS: ^{123}I, Nuclear Medicine, Gas Flow Cyclotron Target, Low-Dose Radiopharmaceutical

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